PRESSED BORIDE CATHODES

W. Wolski

(NASA-TH-77821) FRESSED BOBIL CATHODES (National Aeronautics and Space Administration) 26 p HC A03/MF A01 CSCL 07D N85-27984

Unclas G3/25 21455

Translation of "Prasowane katody borkowe", Elektronika, Vol. 11, No. 4, 1970, pp. 149-155.



NATIONAL AERONAUTICS AND SPACE ADMINISTRATION WASHINGTON, DC 20546 APRIL 1985

NASA TECHNICAL MEMORANDUM

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Results of experimental studies of emission cathodes made from lanthanum, yttrium, and gadolinium hexaborides, from the viewpoint of requirements posed in the production of accelerators. Maximum thermal emission was obtained from lanthanum hexaboride electrodes. The hexaboride cathodes operated stably under conditions of large current density power draw, at high voltages and poor vacuum. A microtron electron gun with a lanthanum hexaboride cathode is described.						
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PRESSED BORIDE CATHODES

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W. Wolski Doswiadczalne Zaklady Lampowe (Experimental Tube Plant)

The term "boride cathodes" signifies cathodes based /149* on diboride and hexaboride metals. In terms of practical application, hexaborides of rare earth metals are especially interesting. These are crystal compounds with a high melting point, considerable hardness and chemical resistance. They exhibit many properties typical of metals, e.g. they are good conductors, their coefficient of thermal resistance is positive, they possess the characteristic metallic luster, etc.

Cathodes based on these materials are characterized above all by high resistance to poison and ion bombardment, high working temperature, low evaporativity, and low efficiency.

The first report on the emission properties of the hexaborides of certain metals was published in 1950 by J. M. Lafferty [17]. In an article the following year [18], he summarized the results of his research, paying special attention to the remarkable emission properties of lanthanum hexaborides. Only cathodes based on barium oxide have a better thermoelectron emission than LaB₆. However, their permissible working temperature is rather low. At high temperatures and under heavy current draw lanthanum hexaboride surpasses all other cathode materials in general use. Comparative studies have shown that, to obtain the same thermal emission, thorium oxide must be heated 150° higher than lanthanum hexaboride [4].

This work studies hexaborides of lanthanum, yttrium, and gadelinium in view of their application in electron accelerators. The research was conducted in the Division of Electron Physics, Royal Institute of Technology, Stockholm [25].

^{*}Numbers in the margin indicate pagination in the foreign text.

Description of boride cathodes

Of the three borides mentioned above, only lanthanum hexaboride has a practical application in electronics. The available literature offers no information on the use of yttrium or gadolinium hexaborides as cathode materials; furthermore, published data characterizing their emission features differ seriously.

Table 1 compares certain physical properties of the borides under discussion, chiefly on the basis of reference [21]. The comparative constant emission values cite' were obtained in specially constructed experimental diodes at relatively low temperatures with constant current supply and pressure below 10^{-7} Tr.

Illustration 1 presents a theoretical set of thermoemission characteristic curves for boride cathodes, as well as several other thermocathodes in general usage. The constant emissions given in table 1 are used to calculate hexaboride characteristic curves.

The chief obstacle to the widescale use of hexaborides as cathode materials is their high activity in relation to practically all metals with a high melting point. Contact between a hexaboride and metal at high temperature results in diffusion of boron to the metal and creates alloys or lesser borides. This causes a weakening of the bonds in the hexaboride crystal structure, lowering its mechanical resistance and facilitating the evaporation of atoms from the metal in the hexaboride compound. The ease of boron diffusion constitutes the chief impediment to employing in lamps directly heated by cathodes coated with a layer of LaB₆ by cataphoresis. For example, a tungsten cathode coated with LaB₆ operated in a temperature of 1515°C only 48 hours [18]. The diffusion of boron to platinum or iridium happens so swiftly that in essence

the cathodes lose their mechanical resistance before attaining the activation temperature [3].

TABLE 1
PHYSICAL PROPERTIES OF RARE EARTH METAL HEXABORIDES

Material	LaB ₆	YB ₆	GdB ₆
Melting point, *C	2530	2300	
Density, g/cm ³	4.72	3.64	5.31
\mathcal{E}_{y} Spectral emission ($\lambda = 0.655 P$ m)	0.70	0.70	0.65
Specific resistance $oldsymbol{\Omega}$ cm	15.10-6	40.10-6	44.7°10 ⁻⁶
Temperature coefficient of resistance	+2.68*10 ⁻³	+1.24.10-3	+1.4°10 ⁻³
Coefficient of thermal expansion	6.4.10-6	6.2.10-6	8.7.10-6
Thermal conductivity W/m°C	47.8	29.4	20.6
Constant Richardson		į	
emissions:Øo , eV	2.60	2.22	2.59
A, $A/cm^2/^{\circ}K^2$	28.8	9.6	9.9
scurce	[22]	[15]	[11]
Secondary emission coefficient [15]	0.95	1.0	0.8



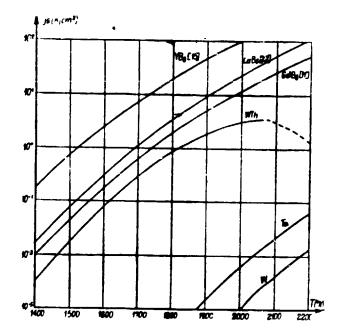


Fig. 1. Thermoelectrode emission of various cathode materials

The difficulties associated with the high activity of borides can be overcome, at least in part, by application of various protective coatings between the core and the boride coating. The function of protective coating can be performed by graphite, boron, or tantalum carbide [18], ThO₂ [1], MoSi₂ [16]. Good results were obtained by employing cathode cores of rhenium [3, 4, 7]. The boride cathode can also be constructed in the form of a rod fastened at one end, with a low working temperature, which significantly reduces the diffusion of boron [2].

The application of a protective coating of MoSi_2 is described in source [16]. The electron gun cathode in the synchrotron is constructed in the form of a tantalum cartridge clip 10 X 1 X 0.5 mm in dimension, with tantalum powder /150 baked onto its outer walls. The porous coating achieved in this manner is impregnated with molybdenum disilicide, and the entire cartridge clip is subsequently filled with a suspension of LaB_6 powder in glue. Experiments with the gun, conducted

in a vacuum of 10^{-5} - 10^{-4} Tr with pulse current consumption at a density of 30 A/cm^2 and a field intensity of 10^7 V/m, revealed that cathode durability totaled approximately 300 hr. At 1650°C it was possible to draw a current with a density of 50 A/cm². Information on the resistance of LaB₆ cathodes to gas poisoning is published in [3, 89, 15]. Cathode poisoning is observed after a certain critical gas pressure is reached, while the poison resistance of the cathode rises with its temperature. The critical pressure for oxygen at 1400°C is $2^{\circ}10^{-6}$ Tr, and at 1570°C, $5^{\circ}10^{-5}$ Tr [8]. A comparison of the resistance of various cathodes at typical working temperatures reveals that the critical pressure of oxygen for LaB is 2-3 orders higher than for oxide or impregnated cathodes. At $1500\,^{\circ}\text{C}$ the critical pressure for atmosphere equals $5^{\circ}10^{-5}$ Tr, for CO_2 -- $2^{\circ}10^{-5}$ Tr, for H_2 , N_2 , and Ar -- over 10^{-2} Tr. The degree of cathode poisoning depends on gas pressure and cathode temperature; for each pressure/temperature there exists a definite and constant emission level. The poisonings are reversible and cathodes can be completely reactivated at 1650°C [15].

From the standpoint of their exceptional resistance to atmospheric influence and their ease of activation, LaB_6 cathodes have found application chiefly in removable vacuum systems, e.g. in charged particle accelerators of various types [5, 9, 12, 16]. Other examples of technical applications are equipment for melting and welding metals, high-powered electron guns, thermionic heat/electricity converters [15, 21], ionization gauges and omegatrons [20], Balzers quadruple mass spectrometers, etc. Experiments were likewise conducted with blade cathodes, the emissions of which are conditioned by the existence of a strong electric field at the cathode surface. A blade cathode constructed of LaB_6 in continuous operation yielded a current of 10^4 - 10^6 A/cm² density over the course of 200 hr [10].

Analysis of literature reveals that cathodes of LaB₆ can in many instances successfully replace so-called high-temperature cathodes, surpassing them above all in emission properties and poison resistance as well as durability under ion bombardment.

Cathode Pressing

Maximum density and concomitant mechanical durability can be attained by employing a method of hot pressing under vacuum conditions [15, 21]. The strong crystal bonds in rare earth metal hexaborides cause the temperatures at which plastic deformation occurs to be very high, near the melting points. This significantly complicates the pressing process. Figure 2 presents schematically the method of hot pressing in a vacuum used in the present work for producing boride cathodes.

The essential parts of the pressing equipment are constructed of graphite, which has sufficient mechanical durability at high temperatures and does not react with borides. Metal hexaboride in powder form is placed in a sleeve between two rollers, and the pressing apparatus is subsequently mounted in a vacuum chamber. The high temperature required for sintering is achieved by heat emission deriving from the flow of current through the apparatus. To exert the necessary pressure, a supplementary weight is attached to the mobile pass pressing on the upper stamp. The pressure applied after many experiments in the present work amounts to 170 kg/cm² for all three types of hexaborides studied. The entire pressing and sintering process takes place in a vacuum better than 10⁻⁴ Tr.

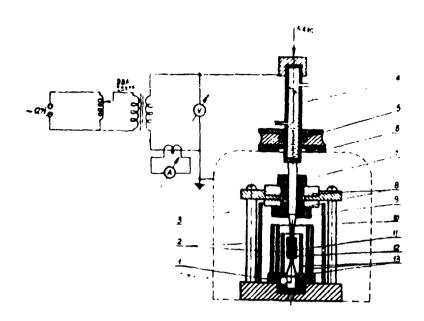


Fig. 2. Hot pressing of hexaborides in a vacuum: 1--stamp, 2--rollers, 3--vacuum tank, 4--water-cooled pass, 5--rubber gasket, 6--teflon plate, 7--graphite cork, 8--ceramic isolator, 9--nut, 10--stamp, 11--hexaboride powder, 12--sleeve, 13--screens

The pressing procedure is monitored by measuring the following parameters: current, temperature of the outer surface of the graphite sleeve, and the descent of the upper pass, which supplies the degree of powder compression.

To illustrate the pressing procedure, figure 3 presents the typical temperature/current function. Temperature is measured by a pyrometer through a glass window in the vacuum chamber and through openings in the screens. While it differs significantly from the actual temperature of the pressed powders, a knowledge of it is useful in determining the pressing parameters.

Current is gradually increased in order to avoid large temperature fluctuations. At maximum sintering temperature,

attained after approximately 40 min, the molded piece is held in place for 20 min. During this time its size decreases by a factor of 2-2.5. The actual temperature of the powder is near melting point, since occasionally melted hexaboride flows out of the sleeve without an observable temperature rise monitored by the pyrometer. Therefore, once maximum temperature is attained, monitoring the pressing procedure in terms of current is more convenient and in practice more reliable than by pyrometer.

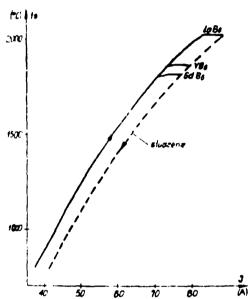


Fig. 3. Relationship of temperature to current. Molded piece diameter 2mm, amount of powder 25-30 mg.

The molded piece cannot be removed from the apparatus without completely destroying the sleeves and graphite rollers. The graphite is partially removed by a knife and then by sandblasting.

The quality of the molded piece's surface depends above all on the production precision of the graphite rollers and sleeves. The rollers must be smooth and accurately fitted to the sleeves in order to prevent the effluence of hexaboride in the event of melting. The molded pieces cannot be machined due

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to their considerable hardness. Their surface can be ground only with a diamond disk.

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The porosity of the cathodes equals sew percent. In the present work only cathodes with a por sw 5% were subjected to emission studies.

Figure 4 presents the cathod construction used in a microtron. An analogous cathod construction was utilized in an experimental system for pulse measurements described below.



Fig. 4. Microtron cathode: 1--LaB₆ tablet, 2--MoSi₂ covering, 3--tantalum retainer

A vacuum-pressed pellet of hexaboride 2 mm in diameter is soldered to a tantalum retainer with the aid of molybdenum disilicide, which simultaneously acts as a coating guarding aginst the diffusion of boron to the tantalum. The melting temperature of MoSi₂ is 2020°C [15].

Boride cathode soldering takes place in a vacuum at a pressure below $5\cdot10^{-4}$ Tr. The soldered joint is covered with a thick suspension of ${\rm MoSi}_2$ in glue. Cathode heating during soldering is accomplished with the aid of an electron gun with a power of 2 kW.

The difference between the coefficients of thermal expansion for hexaborides (table 1) and tantalum $(6.6 \cdot 10^{-6})^{-1}$ is quite small and therefore obtaining a good joint presents no serious difficulties, provided that the width of the gap between the pellet with the hexaboride and the tantalum retainer does not exceed 30 m.

Marie San Carlot

Description of the Method for Measuring Emissions

LaB $_6$ cathodes were studied both under pulse conditions and under constant work conditions; cathodes of YB $_6$ and GdB $_6$ only under pulse conditions. A diagram of the system for pulse measurements is shown in figure 5. A series of electron gun microtron elements were used in the construction of the system.

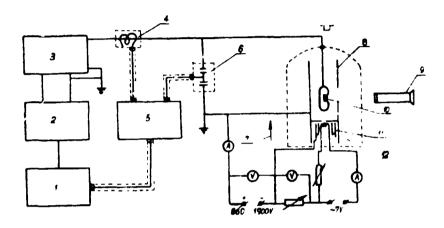
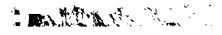


Fig. 5. Diagram of a system for pulse measurements:
1--trigger 12.5-25-50-100 pulses/s, 2--pulser r = 7.2 s,
3--pulse transformer, 4--current transformer 0.1 V/A,
5--oscilloscope, 6--capacitive divider 1:2500,
7--direction of magnetic field, 8--anode, 9--pyrometer,
10--cathode, 11--auxilliary gun

The boride cathode under study is heated by electron bombardment from an auxilliary gun, the directly heated cathode of which is made of tungsten wire. The electron stream is further focused by a magnetic field. An assembly of an anode together with the auxilliary gun, insulated by ceramic material, is installed in a dismountable vacuum station. The cathode under study is placed in the center of the anode on a high tension pass. The relationship of the cathode temperature to the power exerted on it by the auxilliary gun is shown in figure 6.



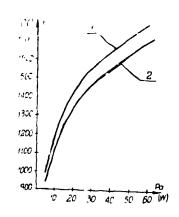


Fig. 6. Relationship of cathode temperature to exerted force 1--actual temperature, 2--temperature as measured by the pyrometer

Cathode temperature is measured by a pyrometer through a glass window in the vacuum chamber, as well as through an opening in the anode. Actual temperature is described by the formula:

$$\frac{1}{T} = \frac{1}{T_S} + \frac{\lambda \cdot \ln \varepsilon_{\lambda}}{C}$$

where:

C -- 1.438 cm 'K

$$\lambda -- 0.655 \cdot 10^{-6} \text{ cm}$$

 $\{\lambda$ -- spectral emission (table 1)

 $T_{\rm S}$ signifies the value of the temperature measured /152 by the pyrometer and increased by a correction for absorption by the glass window. This correction, ascertained experimentally, averages 16° C.

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Negative puls s with a length of 7.2 s and a voltage of up to 60 kV are applied to the cathode. Voltage and current are measured by an oscilloscope. An experimental diode for the study of an LaB₆ cathode under constant work conditions is shown in figure 7. The cathode is heated by an electron gun, as in the pulse method, but without magnetic focusing. The diode is placed in a vacuum station. Because the diameter of the anode opening and the distance between the cathode and the anode are small in comparison to the thickness of the anode, the cathode surface is treated like an absolutely black body in determining the actual temperature, and only a correction for glass absorption is required

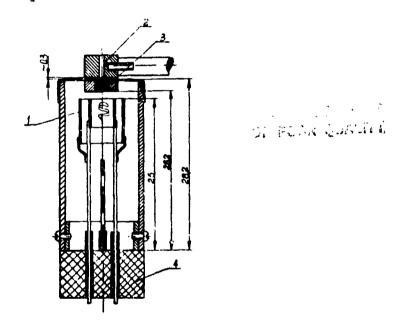
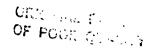


Fig. 7. Diode for measuring emissions under constant work conditions: 1--auxilliary gun, 2--anode, 3--cathode, 4--ceramic isolator

Results of the measurements are analized by Schottky lines, obtained by determining the relationship of lg I to ${\tt U}^{1/2}$ for various temperatures. The current of cathode saturation at a given temperature is obtained by extrapolating the lines to a point corresponding to zero voltage.



Before each series of measurements, the cathode under study is degassed by gradually increasing its temperature to $1650\,^{\circ}\text{C}$ at a pressure below $3\,^{\circ}10^{-5}$ Tr. This treatment suffices for the activation of the cathode. During emission measurements the pressure in the vacuum station equals $8\,^{\circ}$ 10^{-6} -- $3\,^{\circ}10^{-5}$ Tr.

Results of Emission Measurements

Figure 8 presents typical pulse characteristic curves for lanthanum hexaboride cathodes. The high dependence of current on voltage in the saturation zone is noteworthy. At high voltages the characteristic curves exhibit a turning point, beyond which a swift increase in current ensues. At higher temperatures the turning point appears at a lower voltage. The slope of the Schottky characteristic curves increases beyond the turning point, while the saturation current determined at a zero field concomitantly decreases. For these reasons, only those characteristics which correspond to the normal Schottky effect were considered in determining the cathode saturation current.

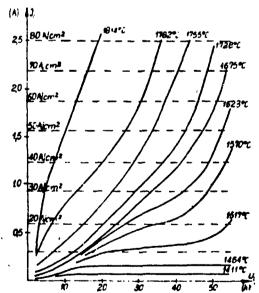


Fig. 8. Pulse current characteristic curve for ${\tt LaB}_6$ cathode.

Under heavy pulse current draw, the cathode emission is frequently unstable. An increase in current at the end of a pulse effects deformation of its shape. It is assumed that the cause is the pulse rise of the anode surface temperature, producing an intense evaporation of anode material, which facilitates a puncture of the inter-electrode region. This hypothesis is supported by observations of microtron operation, in which this phenomenon also occasionally occurs, however it depends on the pulse length and anode construction and material, rather than on the frequency of pulse repetition.

The current characteristic curves of LaB₆ cathodes under constant work conditions are presented in figure 9. Obtaining measurements under higher voltages is hindered by emission of considerable energy in the anode. Thermal radiation from the anode causes cathode temperature instability.

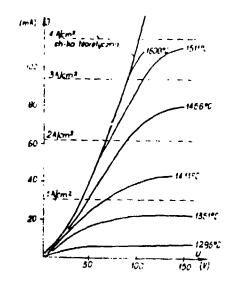


Fig. 9. Characteristic curve of current for LaB₆ cathode under conditions of constant work

The necessity of activating the cathode every time after poisoning appears from an analysis of 1 gures 10. The broken lines represent pulse current characteristic curves /153

obtained immediately after pumping out the vacuum station to a pressure of 3°10 5 Tr. The solid lines represent characteristic curves obtained at these same temperatures, but after

heating the cathode for 10 minutes at 1650°C.

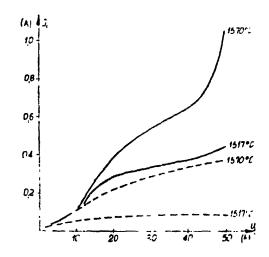


Fig. 10. Influence of subsequent heating at $1650\,^{\circ}\mathrm{C}$ on LaB_{6} cathode emission

Results of incomplete experiments on durability are shown in figure 11. The studies were conducted in a saturation zone with a pulse voltage of 30 kV at 1675°C, which corresponds to 1600°C as measured by the pyrometer. In the course of 120 hr, no deterioration of cathode emissions was observed. Small changes in current were caused by unstable cathode temperatures.

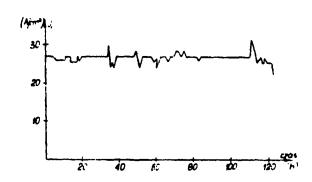


Fig. 11. Durability of LaB₆ cathode.

The current characteristic curves of cathodes constructed of ${\rm YB}_6$ and ${\rm GdB}_6$ are shown in figures 12 and 13.

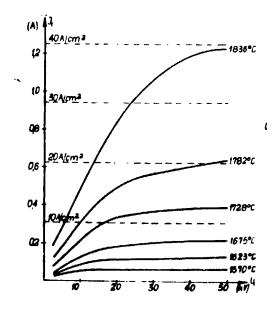


Fig. 12. Pulse current characteristic curve for ${\rm YB}_6$ cathode.

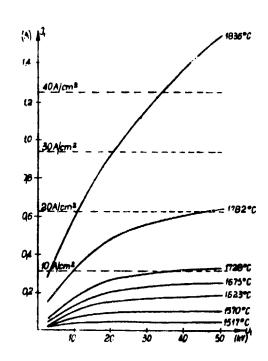


Fig. 13. Pulse current characteristic curve for GdB_6 cathode.

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The emission and behavior of these cathodes are quite similar. The magnitude and shape of the current pulses are quite stable even for high voltages and temperatures. No turning points in the characteristic curves are seen at the saturation zone up to a voltage of 50 kV, and the slope of the curves corresponds to the normal Schottky effect.

Results of emission measurements are compared in table 2. For LaB₆ they agree rather well with the theoretical characteristic curves (figure 1) at a temperature above 1400°C. Poorer emission at lower temperatures should be explained by cathode poisoning from trace gases, as discussed briefly above.

As is to be expected considering the metallic character of borides, pulse emission for lanthanum hexaboride does not differ in essence from emission under constant current supply. The extant discrepancies in the results should be explained chiefly as unavoidable errors in ascertaining actual temperature and in obtaining pulse measurements. While the advantage of lanthanum hexaboride over other compounds is clear, the emission of YB₆ and GdB₆ is nevertheless not bad. It is better for example than the emission of thoriated cathodes. Especially noteworthy is their stability under heavy current dray.

Results of measurements for GdB₆ approximate results of studies published in [11], which were obtained in constant work conditions using experimental diodes with atomized absorbents.

The present research has generally confirmed the good emission properties of boride cathodes under high pulse voltages and relatively poor vacuum, as well as their resistance to multiple poisoning and ease of reactivation.

TABLE 2
RESULTS OF EMISSION MEASUREMENTS

	Thermoelectron emission, $A/cm^{\frac{2}{3}}$				
Temperature	LaB ₆		YB ₆	GdB ₆	
*K	Constant Work				
2109			21.7	15.9	
2055			11.2	11.1	
2001			7.8	6.8	
1948			4.78	4.94	
1896		9.87	3.0	3.66	
1843		6.47	1.82	2.36	
1790		5.88	0.95	55 1.43	
1784	2.38				
1739	1.95				
1737		3.6	0.7	0.685	
1684	0.895	1.97		0.936	
1634	0.535				
1631		0.955	0.4	0.382	
1569	0.189				
1514	$7.54 \cdot 10^{-2}$		}		
1444	2.13 · 10 ⁻²				
1372	5.66 · 10 ⁻³				
1273	$4.22 \cdot 10^{-4}$				
1244	$1.78 \cdot 10^{-5}$				
1214	2.25 · 10 ⁻⁶				
l				1	

Microtron Application of LaB₆ Cathodes

 ${\rm LaB}_6$ pressed cathodes were used in both microtron-type electron accelerators operating in Sweden. The original construction of one is described in [24].

A Comment of the Comm

A microtron is a cyclical accelerator of electrons which pass several times through a resonator gap, thus increasing their kinetic energy. Microwave energy for the resonator is supplied by a 1-MW klystron, at the input of which a low power triode generator operates.

The actual specifications of the microtron operating in the Stockholm Royal Institute of Technology are as follows:

maximum output energy -- 7 MeV

average beam current per pulse -- 0.2 A (for f_p = 12.5 c/s)

resonator frequency -- 2856 MHz

cathode current per pulse -- 3 A

cathode temperature -- 1500-1600°C

maximum electron gun intensity -- 75 kV

pulse length -- 0.1-5 \(\text{p} \) s

Figure 14 presents the construction basis for a microtron gun. A cathode is heated by electron beam bombardment from an auxilliary gun with a tungsten cathode. In comparison to previous constructions this gun is significantly simpler and as characterized by a greater voltage durability. Note that, in an earlier structural version, a pellet of LaB₆ was fastened by means of a rhenium retainer to a tantalum strip directly heated by current flow.

repetition frequency -- 12.5-200 Hz

Lanthanum hexaboride has replaced tungsten cathodes, the low durability of which (on the order of several scores of hours) was the most frequent cause of microtron failure. Similarly, experiments with spongy nickel oxide cathodes met with failure. The durability of these cathodes, due to poor vacuum conditions, scarcely amounted to a few hours. Application of LaB₆ resulted in a significant increase in microtron reliability. At present, cathode durability equals

several thousand hours. The weakest element at present is the auxilliary cathode; due, however, to its lower temperature it can operate approximately several hundred hours and, furthermore, can be replaced in the present structure with little difficulty.

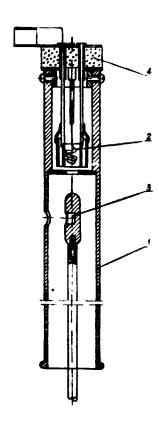


Fig. 14. Microtron gun: 1--anode, 2--cathode, 3--auxilliary gun, 4--ceramic isolator

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